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Abstract

The benchmarking of theoretical modeling is crucial to the ultimate determination of the nature of the electronic structure of Pu. Examples of experimental techniques used for cross checking state of the art calculations will be given.

Introduction

The complexity of the electronic structure of Pu is evidenced by the richness of its binary phase diagram , as shown in Figure 1.

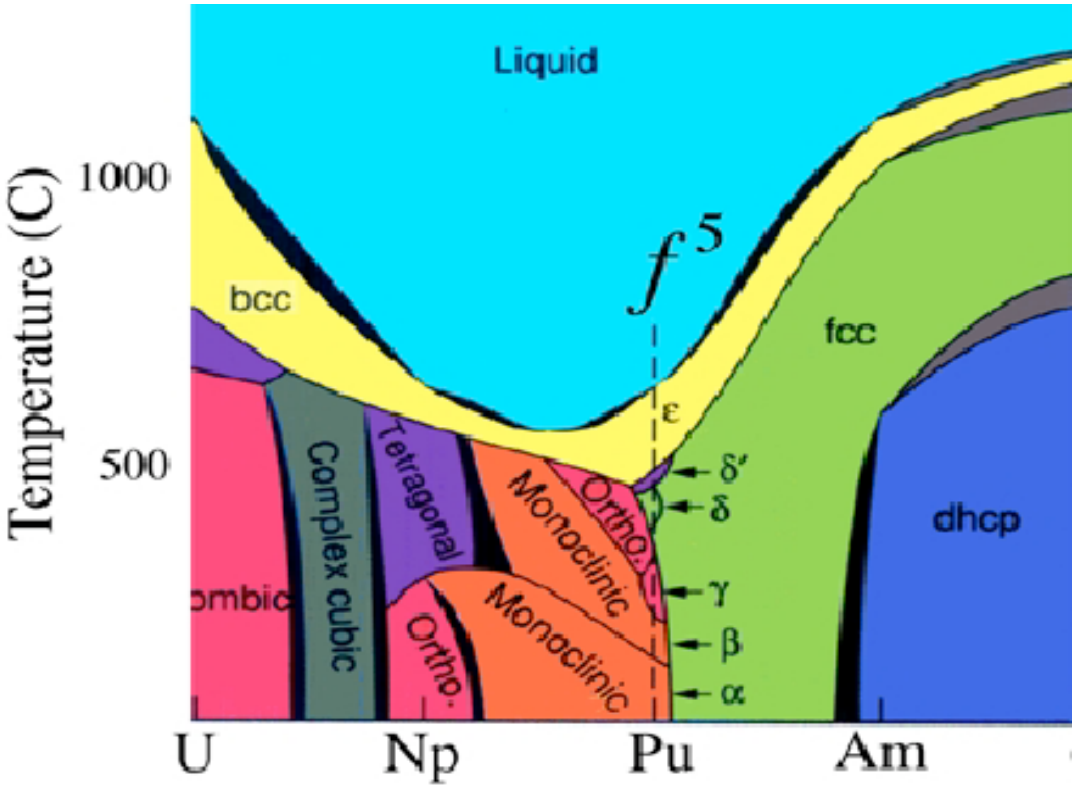


Figure 1.
The actinide binary phase diagram. Taken from Ref 1 and references therein.

In order to benchmark the modeling of Pu electronic structure, several novel experimental approaches have been pursued. The capabilities that supported these measurements will be described here.

Transmission Electron Microscopy at LLNL

The TEM shown in Figure 2 combines capabilities for microscopic imaging, diffraction and spectroscopic analysis via Electron Energy Loss Spectroscopy (EELS). [2,3] In the case of Pu, these capabilities have been applied to achieve a novel result: measurement of the electronic structure of Pu in a phase specific fashion. [4-6] The nano-focusing of the TEM permits the spectroscopic interrogation of small single crystalline of samples in an otherwise polycrystalline sample. The single crystalline nature of these sections can be confirmed directly with electron diffraction. Examples of this type of study are shown in Figure 3.

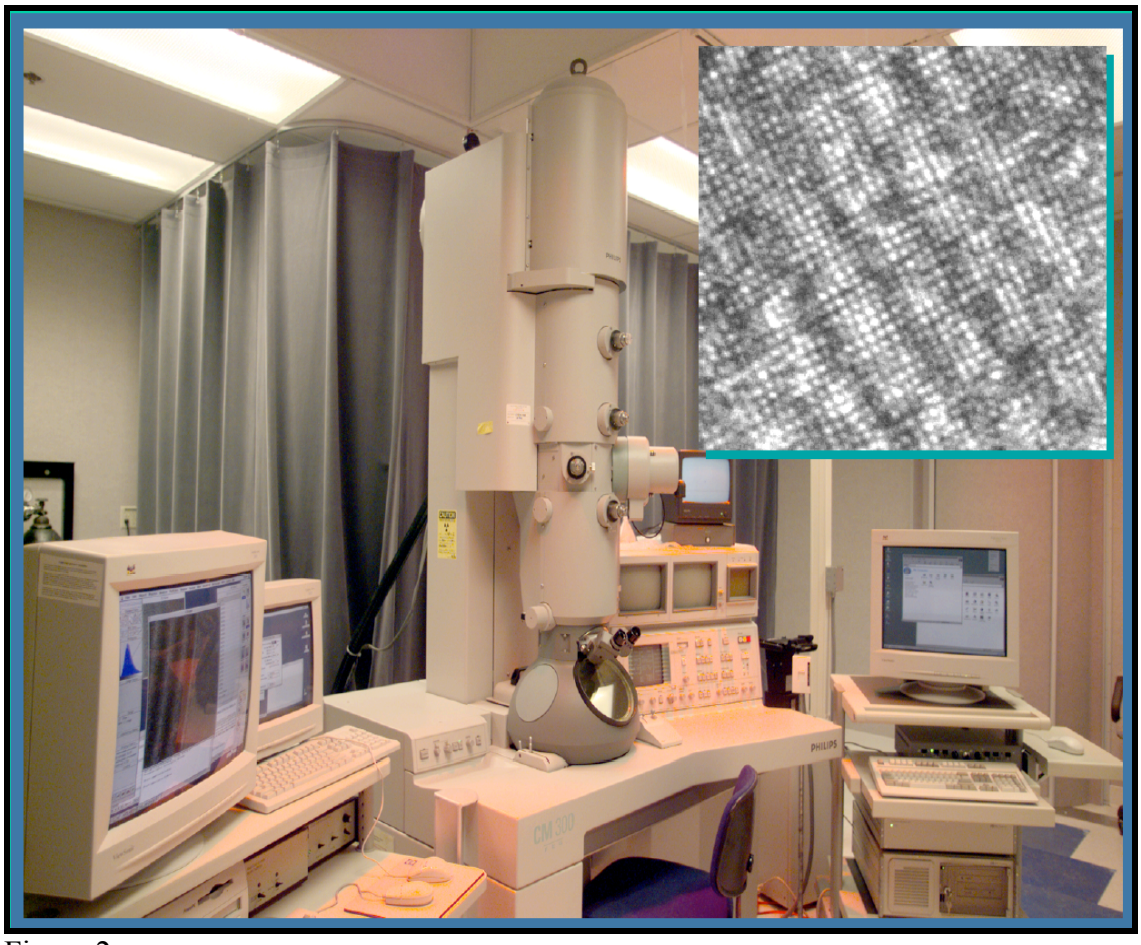
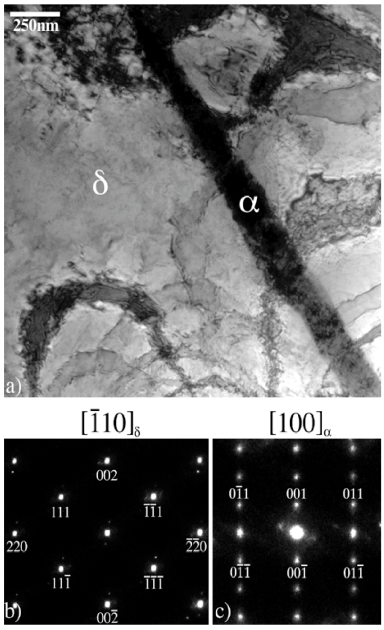


Figure 2
Transmission Electron Microscope (TEM) Facility in the Chemistry and Material Science complex at LLNL. A textured micrograph is shown in the inset.



TEM-Microscopy and HE-EELS

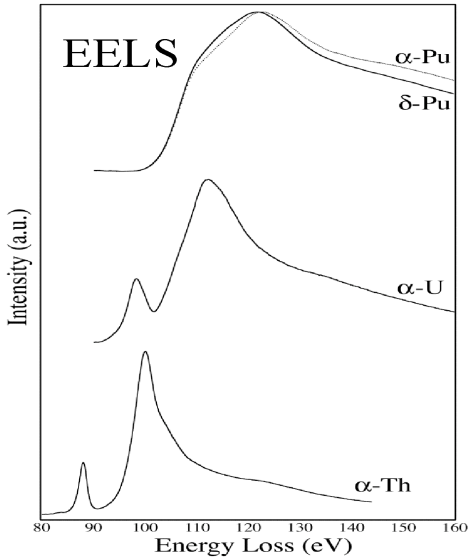


Figure 3
Micrography and diffraction of Pu (to the left) and EELS of Th, U and Pu. [5]

Synchrotron-radiation Based Investigations

Synchrotron radiation from the Advanced Light Source (Figure 4) has been used to investigate the electronic structure of □- Pu and □- Pu. [6,7] The experiments were performed at the Spectromicroscopy Facility (Beamline 7.0) [8]. Measurements include core level and valence band photoelectron spectroscopy, Resonant Photoelectron Spectroscopy (RESPES), and X-ray Absorption Spectroscopy (XAS, Figure 6). [6,7]

Figure 4: Aerial view of the Advanced Light Source at Lawrence Berkeley Laboratory in Berkeley, CA.

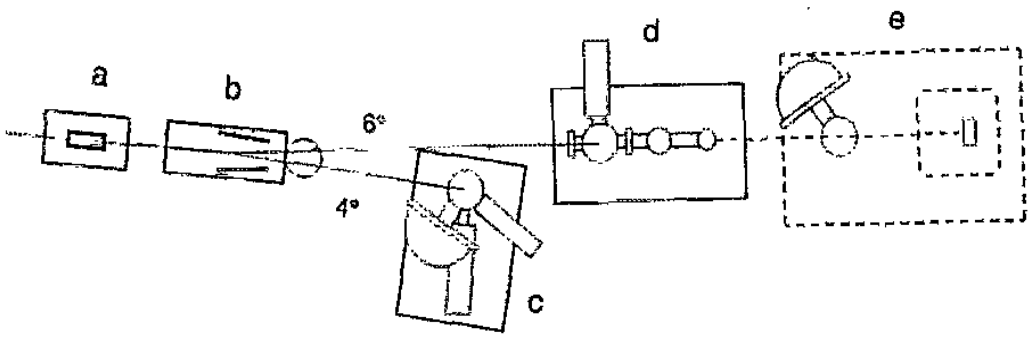


Figure 5
Layout of the SpectroMicroscopy Facility including (a) vertical refocusing optics, (b) horizontal refocusing optics, (c) ultraESCA, (d) x-ray fluorescence and (e) Scanning Photo-Electron Microscope (SPEM). Taken from Reference 8.

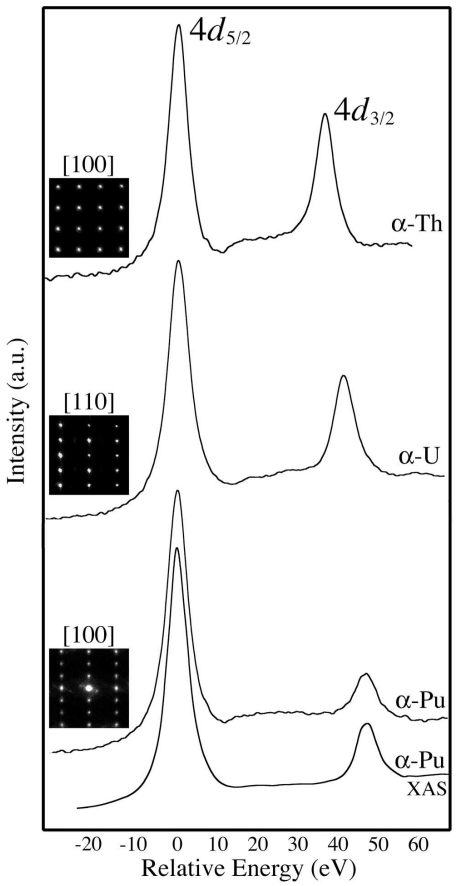


Figure 6
The $N_{4,5}$ (4d -> 5f) peaks from Th, U and Pu acquired by EELS in a TEM and from Pu acquired by XAS. A single crystalline diffraction pattern from each metal is presented, confirming the phase being examined by EELS.

Acknowledgements

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